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LAKE RESOURCES AT RISK TO ACIDIC DEPOSITION IN THE EASTERN UNITED STATES

Jerald L. Schnoor, Sijin Lee, Nikolaos P. Nikolaidis, D.R. Nair
Department of Civil and Environmental Engineering
University of Iowa
Iowa City, IA 52242 USA

ABSTRACT. Watershed descriptors have been obtained or compiled for 1439 watersheds in the northeastern and upper midwestern United States. A methodology, which combines multiple linear regression procedures with a simple deterministic model for alkalinity shows promise as a tool for acid precipitation assessements. Mean absolute errors in predicted lake alkalinity concentrations of approximately ± 100 μeq L^{-1} were obtained with no significant difference (at the 0.05 significance level) between predicted and observed alkalinity histograms. Estimates of the lake resources-at-risk across the depositional gradient from Minnesota to the Adirondack Park of New York were established.

1. INTRODUCTION

The northeastern and upper midwestern portions of the U.S. contain thousands of lakes which are sensitive to acidic deposition, defining sensitivity as acid neutralizing capacity less than 40 μeq L^{-1} measured by Gran titration. The United States Environmental Protection Agency (1985) has recently completed a survey of lakes (greater than 4 ha, less than 200 ha in surface area) and reported about 5% of the Northeastern lakes, and 2% of the Upper Midwest lakes to be acidic. Schnoor et al. (1986) used a simple steady state model to make projections of lake resources at risk to increased deposition in northcentral Wisconsin and the eastern portion of the Upper Peninsula of Michigan. The purpose of this paper is to extend the analysis to a ten state area. Multiple regression statistical analysis based on watershed descriptors for 1439 lakes were combined with a steady state deterministic model to estimate the resources-at-risk at various loadings of acidic deposition.

2. STUDY AREAS

Portions of ten states were included in this study. Moving from west to east, they are: northeastern Minnesota, northcentral Wisconsin, the eastern part of the Upper Peninsula of Michigan, the Adirondack Park of New York, and the New England States of Connecticut, Massachusetts, Rhode Island, New Hampshire, Vermont, and Maine.

Data for the Upper Midwest (Minnesota, Wisconsin, and Michigan) were collected in cooperation with the Wisconsin Department of Natural

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Table I Number and Areal Extent of Lakes in Study Area

Name of Sampled States	No. of Acid Lakes with pH < 5.0	No. of Lakes Sampled	Total Number of Lakes in the Region	Surface Area of Acid Lakes (pH<5.0),ha	Total Surface Area of Lakes in the Region,ha
Adirondacks	208	1,124	2,877	3,786	114,000
Maine	18	752	4,000	335	590,000
Vermont	6	232	525	80	88,000
New Hampshire	17	560	1,088	754	74,000
Rhode Island	8	101	360	94	41,000
Massachusetts	16	624	2,443	260	120,000
Connecticut	1	225	1,218	3	38,000
Michigan (U.P)*	10	53	900	243	25,000
Minnesota (N.E.)*	0	342	2,200	0	100,000
Wisconsin (N.C.)*	18	427	1,600	673	81,000

* The Number of Acid Lakes for Michigan (U.P), Minnesota (N.E.) and Wisconsin (N.C.) was evaluated as Alkalinity < 0.0 ueq/L

Resources (Rhinelander District Office) and the U.S. Environmental Protection Agency (Environmental Research Laboratory-Duluth). The data are compiled in the U.S. EPA "ERLD-UMD Acid Rain Deposition Gradient Susceptibility Database" by Glass (1985). Data for the northeastern states were obtained from several sources including Haines and Akielaszek (1983), ACID database by Hendrey et al. (1983), Pfeiffer and Festa (1980, 1984), Norton et al. (1982) bedrock maps, and the GEOECOLOGY database at Oak Ridge National Laboratory (Olson et al., 1980). These references were compiled into one database at The University of Iowa (UI database).

Table I provides a summary of the number of acid lakes (less than pH 5.0) in each of the study areas. As one moves from west to east, following a gradient of decreasing precipitation pH, the percentage of acid lakes that were sampled increases. In northeastern Minnesota, there were no acid lakes found, while 4.2% were observed in northcentral Wisconsin, followed by 18.9% in the eastern part of the Upper Peninsula of Michigan, and 18.5% in the Adirondack Park of New York. In New England, the percentage of acid lakes sampled ranged from 0.4% in Connecticut to 7.9% in Rhode Island. The number of acid lakes sampled in the combined Upper Midwest and Northeast was less than 7%, based on Table I, and the surface area of these acid lakes represented less than 1% of the total surface area of lakes in the regions.

Since the sampling of lakes in each region was not random nor entirely representative, the percentage of acid lakes is not definitive, but it does illustrate a trend among these study areas. The study areas are all relatively sensitive to acid deposition (crystalline bedrock and/or acidic soils). Among these sensitive regions, the greater is the acidity of precipitation, generally, the greater the percentage of acid lakes. Where the volume-weighted average precipitation pH is 4.6 and the precipitation rate is 100 cm yr^{-1}, the acid deposition is 250 eq ha^{-1} yr^{-1} (wet only). It seems that most lakes in these sensitive areas can generate greater than 250 eq ha^{-1} yr^{-1} of alkalinity to neutralize the acid deposition.

* The Number of Acid Lakes for Michigan (U.P.), Minnesota (N.E.) and Wisconsin (N.C.) was evaluated as Alkalinity < 0.0 ueq/L

3. METHODOLOGY

Utilizing the existing databases, the objectives of this study were to: 1) determine the watershed factors which contribute to lake sensitivity, and 2) predict the number of lakes which would be acidic under various acid deposition loadings.

The UI database consisted of four water quality parameters (alkalinity, pH, conductivity, and color) and 23 watershed descriptors (lake elevation, lake surface area, watershed area, ratio of watershed area to lake area, bedrock type, hydrologic type, number of inlets, number of outlets, drainage order, total watershed relief, number of cabins in the watershed, annual runoff, annual precipitation, annual potential evapotranspiration, annual H-ion loading (eq m^{-2} yr^{-1}), average soil pH, soil percent base saturation, soil cation exchange capacity, and the percentage of the watershed as wooded swamp, marsh, woodlands, open land, or developed land.

Multiple linear regression analyses were used to determine the

watershed variables which correlated well with lake alkalinity in each region. Other investigators have related lake alkalinity to watershed descriptors using multivariate statistics (Rapp et al., 1986; Eilers et al., 1983; Haines et al., 1983; Hunsaker et al., 1985; Baker and Harvey, 1984). The following statistical procedures were utilized in this research.

1) Obtain descriptive statistics (mode, median, mean, kurtosis, and skewness) for each variable in the database.
2) Perform a log-transformation on variables to obtain a near-normal distribution as necessary.
3) Correlate all variables with each other by constructing a matrix of correlation coefficients from linear, least-squares regressions.
4) Use exploratory linear stepwise multiple regression analyses with selected watershed descriptors to determine the best fit (maximum r^2) models with 2-6 independent variables and with alkalinity as the dependent variable.
5) Select the best multiple regression model for each state or region, making sure that the variable associations were logical and reasonably independent. Check the statistical significance for each of the models.
6) Verify the multiple regression model for each state or region by predicting the alkalinity concentrations for lakes using only watershed descriptors and withholding alkalinity data. Estimate the prediction error.
7) Use the steady state deterministic mass balance model to predict alkalinity concentrations under various acid loading conditions.

4. MASS BALANCE MODEL FOR ALKALINITY

The primary drawback to multiple regression models is that they cannot be used to predict alkalinity concentrations under different acid loading scenarios due to the fact that they are dealing with only watershed descriptors. A combination of multiple regression techniques and a deterministic steady state model (Schnoor et al., 1984, 1986; Stumm et al., 1983a) were used to predict the number and areal extent of lake resources-at-risk at double and half the present-day acid loadings. The Adirondack region in New York State was chosen for analysis because the multiple regression model worked best ($r^2 = 0.53$) for this region and because the Adirondack region has the greatest number of acid lakes. The Minnesota and Wisconsin areas were also chosen for analysis because the regression models produced statistically significant predictions of alkalinity based only on watershed variables.

The steady-state alkalinity concentration in the model depends on only four factors: the evapoconcentration factor (I/Q), the amount of precipitation (R), the weathering or chemical neutralization rate (W), and the acidity of precipitation (L_{acy}). W includes all neutralization processes such as chemical weathering, ion exchange, and reduction reactions. The mass balance equation is given below.

$$Alk = \frac{I}{Q}\left\{\frac{W}{R(0.1)} - L_{acy}\right\} \quad (1)$$

where Alk = alkalinity concentration, $\mu eq\ L^{-1}$
I = inflow volume per time, $m^3\ yr^{-1}$
L_{acy} = total acidity of precipitation (dry and wet), $\mu eq\ L^{-1}$
Q = outflow or seepage rate, $m^3\ yr^{-1}$
R = precipitation rate, $cm\ yr^{-1}$
W = weathering and neutralization rate, $eq\ ha^{-1}\ yr^{-1}$
0.1 = units conversion factor.

A mixed kinetic model (with a fractional-order rate and a zero-order rate) has been determined in studies of mineral weathering by Stumm et al. (1983b). The fractional order term represents H^+ attack on mineral surfaces, and the zero-order term represents chemical weathering due to attack of organic ligands. Schnoor et al. (1986) have used an analogous mixed kinetic model to represent neutralization in the watershed, and the following form of the equation is applicable.

$$Alk = \frac{I}{Q}\left\{\frac{k_h\left(\frac{L_{acy}}{L_{ref}}\right)^m + k_o}{R(0.1)} - L_{acy}\right\} \quad (2)$$

where $W = k_h\left(\frac{L_{acy}}{L_{ref}}\right)^m + k_o$ = weathering or neutralization rate

k_h = hydrolysis rate constant for H^+ attack on minerals, $eq\ ha^{-1}\ yr^{-1}$
k_o = ligand weathering rate constant for organic ligands and CO_2, $eq\ ha^{-1}\ yr^{-1}$
L_{ref} = reference precipitation acidity, $\mu eq\ L^{-1}$
m = fractional order weathering rate constant, dimensionless.

One difficulty in the use of the steady state model is how to determine the rate constants, m, k_h, and k_o. Theory dictates that m, the fractional-order hydrolysis rate constant, must be between 0 to 1.0 if the reaction is truly controlled by the rate of dissolution at the surface of minerals. Laboratory results in the literature on acid hydrolysis with a wide variety of minerals indicates that the range of m for most crystalline rock primary minerals is 0 3 to 0.7. A sensitivity analysis was performed as a part of this study, and it indicated that m was best chosen as 0.6 for Adirondacks and Wisconsin, and 0.5 for Minnesota. This optimum value of m for lakes in the three study areas ensured that calibrated rate constants were positive and that alkalinity predictions increased as m increased. Results were not very sensitive to the values of m which were chosen.

Verification of the model (Equation (2)) was performed while withholding measured lake alkalinity field data. Thus, a true prediction error could be obtained. In order to verify the model, a multiple regression equation was used to generate estimated alkalinity and rate constant k_o values from watershed descriptors alone. Then, the model calibration consisted of an inverse procedure to obtain k_h from Equation (2). Finally, Equation (2) was used to predict the alkalinity

concentration of each lake in the data set, and the predicted alkalinity concentration was compared to the measured value.

5. RESULTS AND DISCUSSION

5.1. Regression Models

Linear regression correlations were developed between the alkalinity concentration of the lakes (dependent variable) and each watershed descriptor. The strongest relationships were indicated by correlation coefficients with lake area, lake elevation, annual rainfall, bedrock type, watershed area, soil pH and soil cation exchange capacity (CEC). Lakes in the Northeast (New England states and the Adirondack Park), the Adirondack Park alone, northeastern Minnesota, and northcentral Wisconsin were investigated. A positive correlation coefficient indicates that lake alkalinity increases as the value of the watershed descriptor increases, and a negative correlation coefficient signifies that alkalinity decreases as the watershed descriptor increases. Norton's (1982) bedrock scale was strongly positively correlated with alkalinity of lakes in the Northeast region (r = 0.395) and the Adirondack Park, N.Y. (0.369), while annual rainfall and lake elevation were highly negatively correlated with alkalinity in the Northeast region (-0.403, -0.254), and the Adirondack Park, N.Y. (-0.350, -0.650). In the Midwest region, the bedrock scale (from 1 to 10) of Rapp et al. (1986) was utilized, which goes in the opposite direction of Norton's (1982) bedrock scale. Low numbers on the Rapp et al. (1986) bedrock scale indicate non-sensitive bedrock geology, such as limestone or dolomite. As expected, it correlated negatively with lake alkalinity in Minnesota (-0.262) and Wisconsin (-0.085). The percentage of the immediate watershed area in developed land (agriculture, residential, etc.) was correlated with higher alkalinity concentrations.

In Wisconsin, very strong positive correlations were determined between alkalinity with the lake area (0.450) and hydrologic type (0.723). Hydrologic type was scaled by Eilers et al. (1983) from 1 to 4 according to the potential degree of groundwater influence on the lake. Lakes of hydrologic type 1 are precipitation-dominated seepage lakes, and they represent a class of low alkalinity systems that were clearly discriminated by linear regression analysis. Approximately 50% of the variation in alkalinity among lakes in Wisconsin was explained by the watershed descriptor hydrologic type. H^+ deposition was used as a watershed variable (county-averaged from GEOECOLOGY), and it did not vary enough within a region to be statistically significant in most cases. Figure 1 gives the multiple linear regression equation which was obtained for lakes in the Adirondack Park of New York. The coefficient of determination (r^2) was 0.53, so 53% of the variance in the observed alkalinity concentration values was explained by only three watershed variables (bedrock type, elevation of the lake, and soil CEC). The mulitple linear regression model resulted in an absolute error residual of ± 110 μeq L^{-1} and little bias in prediction (Figure 1).

Figure 1: Plot of Observed vs Predicted Alkalinity Concentration (ueq L^{-1}) for Lakes in the Adirondack Park, New York

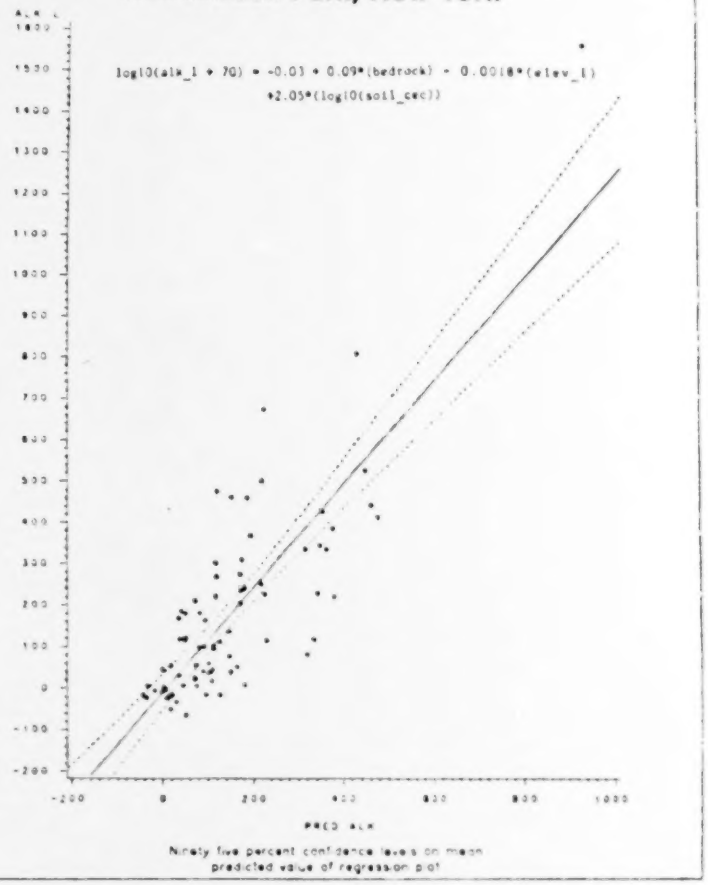


Figure 2: Verification of Trickle-Down Model for Lakes in the Adirondack Park, New York

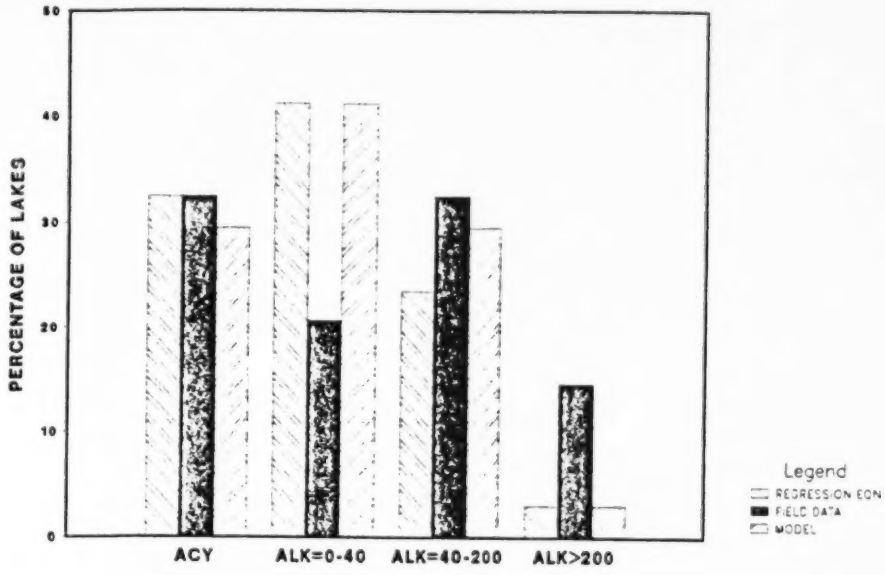
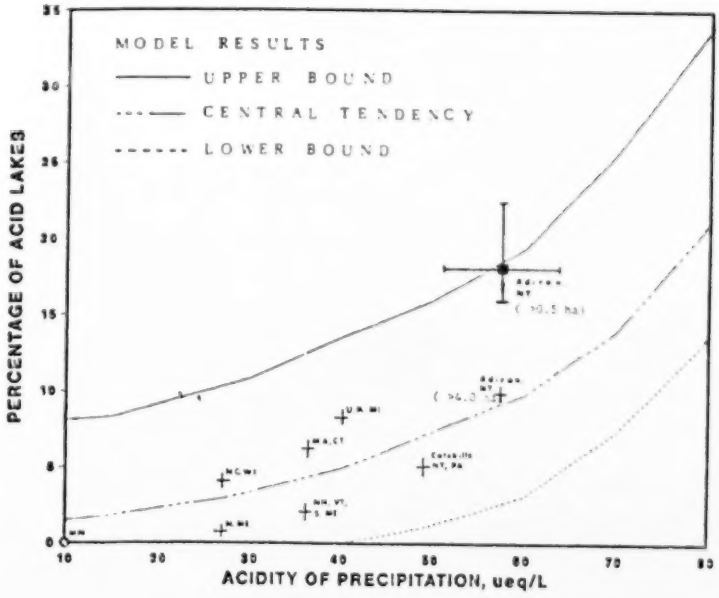


Figure 3: Lake Resources at risk at Varying Deposition



5.2. Steady-State Model

The steady-state deterministic model has sometimes been called the "Trickle-Down Model" by Schnoor et al. (1986) and Stumm et al. (1983a). Results of the model verification are presented in Figure 2 for four alkalinity categories (Alk < 0, 0 ≤ Alk < 40, 40 ≤ Alk < 200, and Alk ≥ 200 μeq L^{-1}). Approximately 32% of the verification set of lakes (n = 34) were measured to be acidic (Alk < 0 μeq L^{-1}), 32% were predicted to be acidic by the multiple linear regression model, and 29% were predicted to be acidic by the steady-state model. The regression equation and the steady-state model gave similar results (which is logical since the steady-state model used the regression equation in its calibration procedure). The models did a poor job of predicting the percentage of lakes in the high alkalinity category (Alk ≥ 200 μeq L^{-1}), but this is not very critical to the risk assessment.

The Steady-state model was used to predict the alkalinity concentration of all lakes in the Adirondack Park at double and half-loading of H^+ deposition. If the loading doubled, the percentage of acid lakes in the Adirondacks would be predicted to increase from 16 to 46%. At half the present acid loading, approximately 6% of the acid lakes in the Adirondacks would recover. These lakes are sensitive to changes, either increases or decreases in acid deposition.

The three lines (upper bound, central tendency, and lower bound) of Figure 3 summarize the results of the study. The crosses indicate the percentage of acid lakes (Gran alkalinity ANC < 0 μeq L^{-1}) in each region according to field data from EPA (1985) on lakes greater than 4.0 ha surface area. The large cross indicated for the Adirondacks includes the range and estimate for all lakes greater than 0.5 ha from this study. The central tendency of all model results is given by the semi-dashed line. In Figure 3, the boundary lines were calculated from Equation (2) using total acidity of precipitation (wet + dry), but the crosses represent available field data based on wet-only precipitation acidity. The central tendency corresponds closely to the model results for northcentral Wisconsin with model coefficients ranging from k_o = 5 - 4778 eq ha^{-1} yr^{-1}, k_h = 144-230 eq ha^{-1} yr^{-1}, m = 0.6, and L_{acy} = 5 μeq L^{-1} background acidity. The solid line corresponds to an upper bound for the most sensitive region, the Adirondack Park of New York, where the model coefficients were k_o = 0 - 7487 eq ha^{-1} yr^{-1}, k_h = 273-613 eq ha^{-1} yr^{-1}, m = 0.6, and L_{acy} = 10 μeq L^{-1}. The lower bound represents the least sensitive region, and it is shown by the dashed line. It corresponds closely to the model results for northeastern Minnesota with model coefficients ranging from k_o = 228-5841 eq ha^{-1} yr^{-1}, k_h = 55-59 eq ha^{-1} yr^{-1}, m = 0.5 and L_{acy} = 1 μeq L^{-1}. The uncertainty of the model results is large, but results are consistent with the field measurements from EPA (1985) and they can be used to estimate (in a preliminary assessment) the lake resources-at-risk to acid deposition.

6. SUMMARY

Multiple regression models and the steady state version of the Trickle-

Down model were used to predict the lake resources at risk under different acid loadings in the Adirondack Park. Error estimates were determined based on a procedure of model calibration, verification (cross-validation), and prediction. A database (UI database) was compiled from existing data sources, including four water quality parameters and 23 watershed descriptors. Watershed descriptors which were significantly correlated with lake alkalinity concentrations were bedrock type, elevation of the lake, annual rainfall, watershed area, lake surface area, and soil pH and CEC.

The Trickle-Down model was coupled with multiple linear regression models. Mean absolute prediction errors were on the order ± 100 μeq L^{-1} alkalinity. The model tended to underpredict the number of high alkalinity lakes (> 200 μeq L^{-1}) and to overpredict the number of lakes in the alkalinity range from 0 to 40 μeq L^{-1} for the Adirondack Park. Application of the Trickle-Down model worked at least as well as the multiple regression equation to predict alkalinity concentrations of the lakes. An estimate of the number of acid lakes in the Adirondack Park under current loading conditions is 16%, but the number would increase to 46% if acid deposition were to double, and it would decrease to 10% at half-loading.

A risk assessment methodology has been developed to estimate the number of acid lakes in any region under current conditions. The prediction error (± 100 μeq L^{-1}) is rather large for an individual lake, but it may be acceptable for estimating distributions of lakes by alkalinity classes. Final verification of the model under different loading conditions is not possible except for cases of whole-lake or whole-watershed manipulation.

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